ATMOSPHERIC BLACK CARBON: CHARACTERIZATION AND DEPOSITION OVER SEMIARID REGION OF INDO-GANGETIC BASIN

A

SYNOPSIS

SUBMITTED FOR THE FULFILMENT OF THE REQUIREMENT FOR THE AWARD OF THE DEGREE OF
DOCTOR OF PHILOSOPHY
IN CHEMISTRY

By

Pratima Gupta

Prof. Sahab Dass
Supervisor
&
Head, Department of Chemistry

Prof. G.S. Tyagi
Dean, Faculty of Science

DAYALBAGH EDUCATIONAL INSTITUTE
(DEEMED UNIVERSITY)
DAYALBAGH, AGRA-282 005 (INDIA)

(2017)

Atmospheric black carbon: characterization and deposition over semiarid region of Indo-Gangetic Basin

1. INTRODUCTION

Black carbon (BC) is a soot particle having heat absorbing nature. It is also known as elemental carbon. It is emitted in the environment during incomplete or partial combustion of fossil fuel, biomass and bio-fuels. It is a complex of light-absorbing mixture and emitted directly into the atmosphere in the form of fine particles ($PM_{2.5}$) (Tiwari et al., 2013). The emitted black carbon can remain in the atmosphere for several days to a week or more. Black carbon is transported from its point of origin to other parts of the globe. The emitted black carbon undergoes vertical distribution and travel from one place to another place according to wind speed and wind direction. It interacts with various components present in the atmosphere before it settling down to the earth's surface via wet and dry deposition (Kumar et al., 2006; Bond et al., 2013). Dry deposition is an important mechanism of removal of pollutants from the atmosphere to the earth surface and affects the surface reflectance. Atmospheric BC alters radiative forcing as it absorbs both incoming as well as outgoing radiations. The sources and effects of BC differ by region to region and season to season. The measurement of black carbon is important for the assessments of exposure and its effects on radiative forcing. The black carbon can be also inferred but the inferential black carbon does not give accurate information. It is inadequate for use in radiative forcing modeling. Hence, measurements of concentration of black carbon should be made at many places. The Asian continent contributes significantly to the world BC emission. The major part of Southeast Asia, Eastern China and the Indo-Gangetic basin of India have been recognized as a hotspot along with equatorial Africa, USA, Mexico and Northern part of Latin America (Ramnathan and Carmichael, 2008). The Indo-Gangetic basin hosts about 40% of Indian populations that are exposed to high load of aerosol particles and has attracted attention of the scientific community worldwide.

However, studies on the measurements of black carbon and its implications for health and radiative forcing are limited.

The term black carbon was first introduced by Tihomir Novakov and he was referred as "the godfather of black carbon studies" by James Hansen, in the 1970s. The ‘smoke’ or ‘soot’ is one of the first known pollutants that have been recognized by the people concerned with the study of deteriorating environment (Hansen, 1970). Soot is composed of a complex mixture of organic compounds which is weakly absorbed in the visible spectral region and a highly absorbing black component which is called as “elemental” or “graphitic” or “black carbon”. The first report of BC aerosol was published in the former USSR in 1967. In the 1970s, however, a continuous study changed the status and demonstrated that black carbon and organic soot components continued to be a large component in urban aerosols across the United States and Europe. Air quality continued to degrade in the developed countries of the world as the population increased. In the late 1970s and early 1980s, surprisingly large amount of black carbon were observed throughout the western Arctic (Rosen et al., 1981). The first measurements of such distributions in the Arctic atmosphere were obtained during 1983 and 1984 as part of the NOAA AGASP (National Oceanic and Atmospheric Administration, Arctic Gas and Aerosol Sampling Program) program with an Aethalometer which had the capability of measuring black carbon on a real-time basis (Hansen et al., 1984). The analysis of this measurement proved heavy deposition of black carbon throughout the western Arctic region thereby showing very high absorption optical depth (Porch et al., 1982). The International Council of Scientific Unions, a coordinating body of national science organizations, launched IGBP in 1986. IGBP (International Geosphere Biosphere Program) aimed to describe and understand how the physical, chemical and biological processes regulate the Earth system. INDOEX (Indian Ocean Experiment) was the first multinational scientific study on chemistry, climate, radiation campaign with multiple platform and single objective. It involves the USA, Europe, India and the Island countries-Maldives, Mauritius and Russia with the primary goal to study radiative and climate forcing by aerosol and feedbacks of regional and global climate.

ARFI (Aerosol Radiative Forcing over India) in India is the core project of Aerosol and Radiative Forcing projects of ISRO-GBP, implemented through SPL (Space Physics Laboratory), Trivandrum. It aims the accurate regional aerosol characterization, incorporating the heterogeneities in space, time and spectral domains for periodical and accurate estimates of Aerosol Radiative Forcing over India and to assess the impacts on regional and global climate.

1.1 **Sources of black carbon**

The major sources of BC are volcanic eruptions, forest fires, etc. along with combustion of coal, biomass, oil, and vehicular emissions (Paliwal et al., 2016). The largest global sources are open burning of forests and savannas, solid fuels burned for cooking and heating and on-road and off-road diesel engines (Sharma et al., 2013). Industrial activities are also significant sources, while aviation and shipping emissions represent a minor contribution to emitted mass at the global scale. Black carbon sources vary by region. In South Asia, the majority of black carbon emission is due to bio-fuel cooking (cow dung cake, coal, wood) while in East Asia, the sources of black carbon emission are coal combustion for residential and industrial purposes (Venkataraman et al., 2005). In Western Europe, traffic seems to be the most important source since high concentrations coincide with proximity to major roads (Dons et al., 2011). Globally, diesel engine accounts for 25 % of all BC emissions and marine vessels also contribute higher levels of black carbon compared to other sources (Jacobason, 2004). The largest contributors of black carbon are Asia, Latin America, and Africa. China and India together account for 25-35% of global black carbon emissions (Streets et al., 2003; Ramanathan and Carmichael, 2008). Ramanathan and Carmichael, 2008 stated that globally 20% of black carbon is emitted from bio fuels burning, 40% from fossil fuels, and 40% from open biomass burning. Global annual emission of BC was estimated to be 8.0 to 24 Tg y^{-1}. Emissions of black carbon increase almost linearly, totaling about 1000 Gg in 1850, 2200 Gg in 1900, 3000 Gg in 1950, and 4400 Gg in 2000 (Bond et al., 2007). Total Asian emission of black carbon was found to be 2.54 Tg, China (1.05 Tg), and India (0.6 Tg) were the first and the

second emitter of black carbon among the Asian countries (Streets et al., 2003). In India, the major contributor of BC (177.4 Gg Y^{-1}) is biomass fuel (Saud et al., 2012).

1.2 **Effects of black carbon**

The presence of BC in atmosphere and its high concentration affects climate and health. BC has become the subject of interest in the recent years due to direct and indirect effects of aerosols on the global radiative balance. They can directly absorb solar and infrared radiation and disturb the energy balance in the earth-atmosphere system (Vaishya et al., 2016). BC aerosols can mix with sulfate, organic carbon, and other water soluble aerosols and become cloud condensation nuclei (CCN) or act as ice nuclei (IN) (Zhang et al., 2011; Tiwari et al., 2013). BC can change the microphysical and radiative properties and lifetime of cloud, and indirectly affect the climate system (Zhang et al., 2011; Vaishya et al., 2016). BC has been associated with a range of climate impacts, including increased temperature and accelerated ice and snow melt. Sensitive regions such as the Arctic and the Himalayas are particularly vulnerable to the warming and melting effects of BC. Black carbon may also be transported to very long distances by wind into areas of high elevation or into the northern latitudes where it deposits onto snow and ice (Zhang et al., 2011). Accumulation of black carbon on snowpack and glaciers can accelerate melting, causing water runoff earlier in the spring and reducing the amount of snowmelt (Thompson, 2003). These effects may further compromise water availability during the drier summer months when it is most needed by humans and ecosystems.

The impact of black carbon on the environment has been increasingly recognized. BC has a short atmospheric lifetime (days to weeks), but strong warming potential. The increased emission of black carbon has also disturbed radiation balance of energy (Babu et al., 2002; Bond et al., 2013). The emission from fossil fuel based industries highly contributes to black carbon or soot which has inherent qualities to absorb the sunlight in significant amount when they particularly lie over the bright surfaces (Herich et al., 2011; Li et al., 2016). They also change the properties of

clouds and indirectly affect the climate (Wang et al., 2011). The impact of black carbon depends upon location and concentration of black carbon in the region (Tiwari et al., 2013). The deposition of black carbon aerosols over highly reflective surface like snow, ice capped peaks and clouds alter their properties by raising their temperature. The decomposition of soot or black carbon over snow or ice directly affect their reflective capacity and results in increasing absorption of heat and in melting of glaciers and polar ice (Clarke et al., 1985; Hansen, 2004). The impact of BC over Indian subcontinent is also visible in quick reduction of glaciers in the Himalayan-Hindu Kush (HHK) region due to 0.25 degree increase in temperature per decade since 1970s (Thompson, 2003).

Black carbon also causes a variety of detrimental effects on human health, agriculture production, plant and the earth's ecosystems (Latha and Badrinath, 2013). Black carbon falls into the smallest size fraction ($PM_{2.5}$) regulated by the US Environmental Protection Agency. These particles can penetrate deep into the lungs where it may enter the bloodstream (USEPA, 2015). Exposure to these particles is associated with respiratory and cardiovascular diseases (Asthma, lung cancer, common cold, heart arrhythmia, stroke, heart failure, etc.) as well as premature death. It is particularly hazardous for people with heart or lung diseases as well as children and old adults (Brook et al., 2010). The urban air quality has reached to such levels that it has started to public breathlessness. Cardiovascular mortality is also associated with black carbon emission. Premature mortality increased due to black carbon exposure in dense traffic and caused inflammatory effect on the respiratory system (Grahame and Schlesinger, 2010; Jansen et al., 2011; Grahame et al., 2014). The exposure of black carbon causes the stress and inflammation marker in the primary school children in a western European urban area (Dons et al., 2014). WHO, 2015 stated that BC, ozone and methane known as short lived pollutant and they contributed significantly to the more than 7 million premature death annually. Black carbon was a better indicator of harmful particulate substances from combustion process (Olstrup et al., 2016).

BC affects the plant growth (reduce the photosynthesis process) and damage agricultural crop (Fowles, 2007). The amount of dust in the atmosphere can have adverse effects carbonyl, hydrocarbons and ozone concentrations (McRae et al., 1994). The increased burning of fossil fuel consumption had changed the scenario of black carbon in the environment (Rhodes et al., 2008). BC also causes the discoloration of building materials (Bergin et al., 2014). Determination of deposition flux of inorganic ions and gases on building material through inferential or parameterization and direct measurements have been reported (Kumar et al., 2002, 2005). The EC have been reported to be responsible for blackening of stone (Brimblecombe et al., 2005). The black carbon deposition on the Taj Mahal surface reported high loading of black carbon influencing the cultural heritage as well as natural and urban surfaces (Bergin et al., 2014).

1.3 Importance of the black carbon study

The variation in the emission source, spatial and temporal distribution, and environmental and climate effects of black carbon are a key area of research. The large uncertainty in global distribution and mixing states of aerosols and knowledge on its direct radiative forcing has made the study of atmospheric BC very important. Today, the majority of black carbon emissions are from developing countries and this trend is expected to increase (Kedia et al., 2014). This results in regional hotspots of atmospheric solar heating due to black carbon. Recently, WHO has released a report in which 13 out of 20 most polluted cities are from India (WHO, 2014). Most of the polluted cities of India are situated over the Indo-Gangetic Basin. It is a major source region for aerosols viz., mineral dust, soot, sulfate and organic particles (Srivastava et al., 2012; Kumar and Kumari et al., 2015; Latha et al., 2016). The IGB region has various types of emission sources. In rural areas, bio-fuels burning (wood, cow dung cake and crop waste) contribute to the high loading of black carbon (Habib et al., 2006) while in urban areas dominant source of the emission of black carbon are fossil fuel burning (coal, petrol, diesel, etc.) (Tiwari et al., 2009; Ram and Sarin, 2010). Agra is

one of the most polluted cities in India over the Indo-Gangetic basin. It is also famous all over the world as it hosts the 'Taj Mahal' one of the seven wonders in the world and other monuments. The high load of aerosol may be due to excessive vehicular emissions, open burning and road paved dust. Hence, it is expected that there may be high loading of black carbon also.

2. REVIEW OF LITERATURE

Scientific community has paid attention on the study of black carbon due to the significant impact of BC on radiative forcing, climate change and its effects on human health as well as on plants and ecosystem. A number of studies have been reported on measurements of black carbon mass concentration, its raditive forcing and climate change effects as well as its effects on ecosystem in India and abroad. However, black carbon study is in primitive stage and sufficient datasets are not available.

2.1 BC mass concentration

National status

The average BC mass concentrations obtained using an aethalometer during January-March 1999 at Goa, a coastal station bounded by Arabian Sea on its west and Western Ghats on its East was about 3 μg m^{-3} (Leon et al., 2001). Extensive ground based measurements of aerosol Black Carbon (BC) and size segregated aerosol mass concentrations have been reported from tropical coastal station, Trivandrum. BC has shown diurnal (with a nocturnal peak and mid afternoon low) and seasonal variations (highest during winter and lowest during monsoon). Daily mean BC concentration as high as ~6 μg m^{-3} was observed on several occasions (Babu et al., 2002). BC mass concentrations measured using an aethalometer in Bangalore, a continental urban location were found to be in the range of 0.4-10.2 μg m^{-3} during November 2001 (Babu et al., 2002). The mean BC mass concentrations over Mumbai, an urban, densely populated and

industrialized city on the west coast of India, during January-March 1999 was 12.4 μg m^{-3} (Venkataraman et al., 2002). Latha and Badarinath, 2003 have made measurements of black carbon aerosols at traffic density to see the impact of mobile sources on black carbon concentrations at Hyderabad, India and found high concentration of BC by a factor of ~2 during morning (0600 to 0900 h) and evening hours (1900 to 2300 h) compared to afternoon hours. Moorthy et al., 2004 reported near surface BC mass concentration of 3.5 μg m^{-3} over Hyderabad. Jayaraman et al., 2006 have reported the BC mass concentrations measurement in western and central India employing the same aethalometer used in the current study. The average BC mass concentration during the campaign was about 2 μg m^{-3}. Kanpur is an urban location in northern India where BC concentrations were found to be in the 6-20 μg m^{-3} range during December 2004 (Tripathi et al., 2005). Black carbon (BC) aerosols were monitored continuously at Pune, a tropical urban location in southwest India. BC concentrations were maximum (about 80% more than annual mean) in winter mainly due to prevailing meteorological conditions like low wind speeds and low ventilation coefficients; as well as due to the transport from northeast regions (Safai et al., 2007.) Badrinath et al. (2007) have reported the BC mass concentration in the range of 15 μg m^{-3} to 50 μg m^{-3} at Allahabad during December 2004. Baxla et al. (2009) have reported seasonal and diurnal behavior of atmospheric submicron aerosol in the northern Indian city of Kanpur in the Indo-Gangetic Plain. The diurnal variation of BC and submicron aerosol mass followed very similar trends in all the seasons. Badarinath et al. (2009) have studied the black carbon (BC) aerosol mass concentration variation in contrasting environments of India in urban and rural areas and reported considerable contrast in its spatial variations. Pathak et al., 2010 have reported the mass concentration of black carbon (BC) aerosols over Dibrugarh in Northeast India for the period June 2008 to May 2009. The BC mass concentration is highest (16.3 μg m^{-3}) in winter and lowest (3.4 μg m^{-3}) in monsoon. Rehman et al., 2011 have studied BC concentrations during cooking hours, both indoors and outdoors. The concentrations of BC reaching 60 μg m^{-3} in the morning and in the evening cooking hours. Reddy et al., 2012 have studied the BC mass concentration over the Anantpur during August

2006 to July 2007. Safai et al., 2012 have reported vertical profiles of black carbon (BC) aerosol and determined from aircraft measurements under the Cloud Aerosol Interaction and Precipitation Enhancement Experiment (CAIPEEX) program during 2009 over Bangalore and Hyderabad. BC mass loadings decreased approximately from 103 to 104 ng m^{-3} at the surface to ~102 ng m^{-3} at an altitude of about 7 km. Kant et al., 2012 have reported Ground based measurement of black carbon (BC) over Dehradun during January to December 2007. The annual average BC concentration was found to be 4.3 μg m^{-3}. The diurnal variation of BC shows a gradual build up in the morning hours between 0600 to 0900 local time and in evening from 1900 to 2200 hrs local time while low concentration is observed during day and night time. Kompalli et al., 2013 have indicated significant BC mass concentration (6.5 μg m^{-3}) at Kharagpur during 2004-2008. Singh et al., 2014 have studied the BC mass concentration in Patiala during October 2008-May 2009. The variation of BC mass concentration has been found in the range of 2 μg m^{-3} to 20 μg m^{-3}. Mahapatra et al. (2013) have studied black carbon (BC) mass concentration variation over Bhubaneswar. Daily, monthly and seasonal measurements revealed a clear winter maxima (5.6 μg m^{-3}) of BC followed by post-monsoon (4.05 μg m^{-3}), monsoon (3.02 μg m^{-3}) and pre-monsoon (2.46 μg m^{-3}). Kompalli et al., 2014 have quantified the role of atmospheric boundary layer in producing temporal changes in BC. The seasonal variations in BC concentrations have a strong inverse relationship with mixed layer height and ventilation coefficient. The rainfall is scanty or insignificantly small during the dry season. Joshi et al., 2015 have made long-term (2009-2012) ground-based measurements of aerosol black carbon (BC) from a semi-urban site, Pantnagar in the Indo-Gangetic Plain (IGP) near the Himalayan foothills. Sarkar et al., 2015 have reported a continuous monitoring of black carbon (BC) aerosols over a high altitude station Darjeeling at eastern part of Himalaya in India during January 2010-December 2011. BC concentration varied between 0.2–12.8 μg m^{-3} with an average of 3.4 μg m^{-3} over the entire period of study. Raatikainen et al., 2015 have conducted the measurements of BC in northern India at two sites: Gual Pahari located at the Indo-Gangetic plains (IGP) and Mukteshwar at the Himalayan foothills. Vaishya et al., 2016 have reported two years of

aerosol spectral light absorption measurements, using filter based technique, from the central Indo-Gangetic plain (IGP), Gorakhpur. Spectral absorption analyses were 4 time higher in winter (W) and post-monsoon (PoM) seasons at UV wavelengths as compared to IR wavelengths. Aerosols from the biomass sources contribute ~28%. Kompalli et al., 2016 have reported the BC mass concentration in Nagpur. Li et al., 2016 have reported that combustion-derived black carbon (BC) aerosols accelerate glacier melting in the Himalayas and in Tibetan Plateau (the Third Pole).

International status

Bodhaine, 1995 have studied the mean BC mass concentrations measured using aethalometers over Barrow, Alaska and Mauna Loa, Hawaii during 1988-1993 and concentration were less than 1 μg m^{-3}. BC mass concentrations measured using an aethalometer at Jungfraujoch a high-altitude station in Switzerland during June 1995 to June 1997 were found to be in range of 0.1-0.8 μg m^{-3} (Lavanchy et al., 1999). Allen et al. (1999) have reported BC in Uniontown, Philadelphia, USA during the summer of 1990 found diurnal and weekly temporal variations. Daily averaged BC mass concentrations were found to be lowest at 0.65-1.49 μg m^{-3}. Kim et al. (1999) have studied the EC mass concentrations in Cheju, an island station. EC mass concentrations were in the range of 4.86-9.86 μg m^{-3}. Im et al. (2001) have measured black carbon concentrations using an aethalometer at Mount Gibbes in the southeastern United States. BC mass concentrations in the polluted continental air masses were about 0.3 μg m^{-3} during summer which reduced to about one third in winter. He et al., 2001 have reported the average BC mass concentrations to be 8.7-10.1 μg m^{-3} at Chegongzhuang, Beijing. The average BC concentrations at Saint Denis, an urban site of La Re´unionisland in the Indian Ocean in November 1996 were found to be in the range of 0.27-0.65 μg m^{-3} (Bhugwant et al., 2000). The BC mass concentrations over Maldives were found to be high and in the 2.7-9.4 μg m^{-3} (Chowdhury et al., 2001). Ruellan and Cachier, 2001 have reported the mean BC mass concentrations during August-October 1997 in Paris nearby a high traffic road at about 14 μg m^{-3}. BC mass concentration measurements were made at East St. Louis an urban site in

Illinois (Sullivan et al., 2004). Byčenkienė et al., 2011 have made continuous measurements of black carbon (BC) aerosol mass concentration at a background site Preila during the period 2008-2009. Monthly means of BC concentration ranged from 212 to 1268 ng m^{-3} and the highest hourly means of concentration were from 4800 to 6300 ng m^{-3}, predominantly in spring and winter months. Chen et al., 2014 have reported BC mass concentration at an urban site of Guangzhou in South China which ranged from 0.6 to 20.5 μg m^{-3}, with an average value of 4.7 μg m^{-3}. Cao et al., 2014 have studied the optical properties of BC, such as depolarization ratio, extinction coefficient, optical depth, Ångström exponent and effective radius. Kanaya et al., 2016 have made long-term (2009-2015) observations of atmospheric black carbon (BC) mass concentrations using a continuous soot-monitoring system (COSMOS) at Fukue Island, western Japan and provided information on wet removal rate constraints and the emission strengths of important source regions in East Asia (China). The BC concentration has been reported to be four times higher than concentration on city sidewalk (Jeong et al., 2017).

2.2 Radiative Forcing

National status

Babu et al., 2002 have organized a comprehensive aerosol field campaign in which simultaneous measurements were made of aerosol spectral optical depths, black carbon mass concentration and size segregated aerosol mass concentrations over an urban continental location Bangalore. Large amounts of BC were observed; both in absolute terms and fraction of total mass (~11%) and sub-micron mass (~23%) implying a significantly low single scatter albedo. The aerosol visible optical depth was in the range of 0.24 to 0.45. Estimated surface forcing was as high as -23 W m^{-2} and top of the atmosphere (TOA) forcing was +5 W m^{-2} during relatively cleaner periods. The net atmospheric absorption translates to an atmospheric heating of ~0.8 K day^{-1} for cleaner periods and ~1.5 K day^{-1} for less cleaner periods. Dey et al., 2003 have retrieved the aerosol

parameters over the Arabian Sea and the Bay of Bengal. Top of the Atmosphere (TOA) and the surface forcing have been deduced over the Arabian Sea and the Bay of Bengal. In general, higher forcing over the atmosphere has been observed over the Bay of Bengal (30 to 54 W m^{-2}) compared to those over the Arabian Sea (26 to 48 W m^{-2}). Tripathi et al., 2005 have made extensive measurements of aerosol black carbon during December 2004 at Kanpur as a part of ISRO-GBP comprehensive aerosol land campaign. The estimated surface forcing is as high as 62 ± 23 W m^{-2} and top of the atmosphere (TOA) forcing is +9 W m^{-2}, which means the atmospheric absorption is +71 W m^{-2}. Panicker et al., 2010 have discussed the extent of Black Carbon (BC) radiative forcing in the total aerosol atmospheric radiative forcing over Pune. The atmospheric radiative forcing for composite aerosols were found to be +35.5, +32.9 and +47.6 Wm^{-2} during post-monsoon, winter and pre-monsoon seasons, respectively. The average BC mass fraction was found to be 4.83, 6.33 and 4 µg m^{-3}during the above seasons contributing around 2.2 to 5.8% to the total aerosol load. Panicker et al., 2008 have reported surface forcing of 9.4 W m^{-2}. Study reveals that about 25% of the aerosol shortwave cooling is being compensated by increase in long wave radiation due to aerosol absorption. Verma et al., 2011 have studied the aerosol radiative effects due to aerosols emitted from different emission sectors (anthropogenic and natural) and originating from different geographical regions within and outside India during the northeast (NE) Indian winter monsoon (January-March). Aerosols lead to the reduction in surface radiation by 40-60% of the total reduction in surface radiation due to all aerosols over the Indian subcontinent and adjoining ocean. Das and Jayaraman, 2011have studied and addresses the role of black carbon (BC) in aerosol radiative forcing (ARFI) over western Ahmedabad (urban area), Udaipur (semi-arid region) and Mt. Abu (a hilltop representing background conditions), India. Surendran et al., 2013 have reported the radiative effects of black carbon (BC) aerosols over New Delhi using Santa Barbara DISTORT Atmospheric Radiative Transfer (SBDART) model. Kaskaoutis et al., 2013 have reported the aerosol episode (AE) days and examines the modification in aerosol properties and radiative forcing during the period 2001-2010 based on Kanpur-AERONET data. Raju et al., 2015 have

reported that the presence of BC resulted in positive radiative forcing in the atmosphere leading to warming effect (+ 2.1 W m^{-2}) whereas cooling was observed at the top of the atmosphere (− 0.4 W m^{-2}) and at surface (− 2.5 W m^{-2}). Using the OPAC and SBDART models, optical properties and aerosol radiative forcing (ARF) in the spectral range 0.2 to 4 μm for composite aerosol and without-BC aerosol at the top of the atmosphere, surface and atmosphere were computed. Patel and Kumar, 2015 have reported during pre-monsoon 2012 using ground measurements, satellite observations and model simulations. The mean top of the atmosphere (TOA) and surface forcing come out to be -14.49 W $m^{-2}$and -53.29 W m^{-2}, respectively. The mean net atmospheric radiative forcing was 38.79 W m^{-2}.Latha et al., 2016 have studied black carbon radiative effects at the densely populated plain station, Varanasi and the lesser populated plateau station Ranchi with large forest cover but with numerous open coal mines.

International status

Aerosol affects the Earth's temperature and climate by altering the radiative properties of the atmosphere (Jacobson, 2001). Yu et al., 2001 have made an effort to reduce uncertainties in the quantification of aerosol direct radiative forcing in the southeastern United States (US). The cloud-free instantaneous TOA ADRFs for highly polluted, marine and continental air masses range from -20.3 to -24.8, -1.3 to -10.4, and -1.9 to -13.4 W m^{-2}, respectively. Jacobson (2004) have reported that aerosols affect the Earth's temperature and climate by altering the radiative properties of the atmosphere. This finding implies a higher positive forcing from black carbon than previously thought, suggesting that the warming effect from black carbon may nearly balance the net cooling effect of other anthropogenic aerosol constituents. Alam et al., 2011 have analyzed aerosol optical properties through the ground-based Aerosol Robotic Network (AERONET) over the mega city Karachi during August 2006–July 2007. The aerosol radiative forcing (ARF) for the whole observation period at the top of the atmosphere (TOA) is in the range of -7 to -35 Wm^{-2} at the surface from -56 to -96 W m^{-2} increasing the atmospheric forcing from +38 to +61 W m^{-2} (average

+51±13 W m^{-2}). Bond et al., 2013 have studied global atmospheric absorption attributable to black carbon. The best estimate for the industrial era (1750 to 2005) direct radiative forcing of atmospheric black carbon is +0.71 W m^{-2} with 90% uncertainty bounds of (+0.08, +1.27) W m^{-2}. Wang et al., 2014 have reported the radiative forcing and climate response due to black carbon (BC) in snow and/or ice. The results show that the global annual mean surface radiative forcing due to BC in snow/ice were +0.042 W m^{-2}, with maximum forcing found over the Tibetan Plateau and regional mean forcing exceeding +2.8 W m^{-2}. Jian et al., 2004 have found that BC aerosols induce a positive radiative forcing at the top of the atmosphere (TOA), which is dominated by shortwave radiative forcing. The maximum radiative forcing occurs in North China in July and in South China in April.

2.3 Dry deposition

National status

A limited study on dry deposition has been carried out. Suman, 2007 have reported the dry deposition monitoring throughout central Panama in 1981 in order to collect particulates produced during the period of agricultural burning. Latha et al., 2005 have reported the estimation of black carbon aerosol scavenging coefficient by using ground-based measurements over an urban environment of Hyderabad during January to December 2004. Kumar and Kumari, 2012 reported the parameterization method of dry deposition of S compounds on natural surface (leaf of *Cassia siamea*) through direct measurement method. Nair et al., 2013 have studied the absorbing aerosols such as black carbon (BC) or dust over high-altitude Himalayan regions (Hanle and Nepal Climate Observatory-Pyramid) have potential implications on the regional climate and hydrological cycle over South Asia. The estimated amount of BC in the snow varied from 117 to 1.7 µg kg^{-1} for a wide range of dry deposition velocities (0.01-0.054 cm s^{-1}) of BC, snow depth (2-10 cm) and snow densities (195-512 kg m^{-3}). Reddy et al., 2000 have used box model to compare the burdens, optical

depths and direct radiative forcing from anthropogenic $PM_{2.5}$ aerosol constituents over the Indian subcontinent. The contribution from various aerosol constituents was 28% sulphate, 25% mineral (clay), 23% fly-ash, 20% organic matter and 4% black carbon.

International status

Koch and Hansen, 2005 have studied the impact of black carbon (BC) on the sensitive Arctic climate, possibly altering the temperature profile, cloud temperature. The GEM-AQ model suggested that the below-cloud scavenging dominates the contribution of BC removal over the Arctic with an estimation of 48% for 2001, whereas the contributions of in-cloud scavenging and dry deposition were about 27% and 25%, respectively. Takeda et al., 2012 have performed quick measurements of black carbon (BC) particles deposited on foliar surfaces of forest tree. An optical method was used for measuring the amount of BC extracted from foliar surfaces and collected on quartz fiber filters. The absorption spectrum of the filters was measured by spectrophotometer with an integrating sphere. Hienola et al., 2013 have reported the regional aerosol-climate model and suggests that the excessive wet removal is not the main cause of the low black carbon concentration output. Ruppel et al., 2013 have reported the spheroidal carbonaceous particles (SCP) are a well-defined fraction of black carbon (BC). Generally, trends of SCP deposition and model results showed the rapidly increasing values in the year of 1950-1980, and a decrease towards the present. Ginot et al., 2014 have studied a shallow ice core at the summit of Mera Peak in the southern flank of the Nepalese Himalaya range. The analyses revealed that mass fluxes were a few orders of magnitude higher for dust (10.4 ± 2.8 g m^{-2} yr^{-1}) than for refractory black carbon (rBC) (7.9 ± 2.8 mg m^{-2} yr^{-1}). The fraction of dry deposition to total deposition increases from 16% to 25%. Yasunari et al, 2013 have reported BC dry deposition rates in a wide range of 270-4700 μg m^{-2} during the study period. Huang et al., 2014 reported the anthropogenic aerosol components in the Arctic troposphere, such as black carbon (BC) and showed a strong seasonal variation

characterized by a peak in later winter and early spring. The estimated global BC burden is 0.28 Tg. Hadley, 2010 have reported modeling studies of snow and ice by black carbon deposition. This study provides one of the first direct measurements for the efficient removal of black carbon from the atmosphere by snow and its subsequent deposition to the snow packs of California. Yasunari et al., 2010 have reported the possible minimal range of reduction in snow surface albedo due to dry deposition of black carbon (BC).

2.4 BC Inventory

National status

Sharma et al. (2013) have reported that India is known to emit large amounts of black carbon (BC) particles, and the existing estimates of the BC emission from the transport sector in the country widely range from 72 ~ 456 Gg/year (for the 2000's). Gadhavi, 2015 have evaluated emission inventories of black carbon (BC) using Lagrangian particle dispersion model simulations and BC observations from a rural site in southern India. Burkhart et al., 2015 have studied a new GIS based comprehensive high resolution grid wise emission inventory of Black carbon. Domestic fuels contributed maximum (48%) followed by Industry (22%), Transport (17%), open burning (12%) and others (1%). Paliwal et al., 2016 have reported black Carbon (BC) emissions from India for the year 2011 were estimated to be 892.12 ± 151.53 Gg/yr.

International status

Koch and Hansen, 2005 have studied black carbon (BC) particles, derived from incomplete combustion of fossil fuels and biomass and found to have a severe impact on the sensitive Arctic climate, possibly altering the temperature profile, cloud. Cao, 2006 have reported the emission inventories of black carbon (BC) and organic carbon (OC) from China. The emissions were 1500 Gg for BC and 4100 Gg for OC which is mainly due to the burning of coal and bio fuels. Bond et

al., 2007 have reported an emission inventory of primary black carbon (BC) and primary organic carbon (OC) aerosols from fossil fuel and bio fuel combustion between 1850 and 2000. Emissions of black carbon increase almost linearly, totaling about 1000 Gg in 1850, 2200 Gg in 1900, 3000 Gg in 1950, and 4400 Gg in 2000. Wang, 2011 have studied BC emission inventories for North America, Europe, and Asia. Núñez, 2014 have studied a black carbon (BC) emission inventory for Mexico. Chow, 2010 have reported the particulate black or elemental carbon (EC) (black carbon (BC). Chowa, 2006 have reported emission inventories for black or elemental (BC or EC) and organic (OC) carbon can be derived by multiplying $PM_{2.5}$ emission estimates by mass fractions of these species in representative source profiles. The total estimated EC emissions of 432 Gg yr^{-1} was apportioned as 42.5% from biomass burning, 35.4% from non road mobile sources, 15% from on road mobile sources, 5.4% from fossil fuel (e.g., coal, oil, and natural gas) combustion in stationary sources, 1% from other stationary industrial sources, and 0.5% from fugitive dust. Annual BC emissions had increased from 5.3 to 9.1 tera grams during this period. BC emission intensities had increased in the industrial and agricultural sectors, mainly due to an expansion of low-efficiency industry (coke and brick production) in developing countries and to an increasing usage of diesel in agriculture in developed countries.

OBJECTIVES

Black carbon is the second most global warming causing element and is responsible for half of the total current warming impact. The human generated BC emissions are contributing largely to ongoing climate change including global warming. India is reported to be one of the top emitters of BC in the world, whereas the Indo-Gangetic basin tops in the country. Agra is also a populated site in the Indo-Gangetic basin and suffers from high load of air pollution. However, a very few detailed real time black carbon study at Indo-Gangetic basin in general and at Agra in particular have been done and it is high time that such a study is systematically undertaken at Agra. The average life span of BC is from a few days to weeks but it plays a significant role in atmospheric warming and affects human health, vegetation and materials. It is basically minute in size and mostly remains suspended in the atmosphere, and settle onto the surface via dry and wet deposition. Dry deposition is an important mechanism of removal of pollutants from the atmosphere to the earth surface in India as dry conditions prevails for most part of the year while rains are confined to a short monsoon period. However, study on dry deposition of black carbon is limited and none from India as it is a cumbersome method. With this in view the objectives of the present investigation are:

- to measure the concentration of black carbon in ambient air at Agra
- to examine seasonal variation in the concentration of black carbon
- to measure and estimate the dry deposition flux of black carbon,
- to study the seasonal variation in dry deposition flux of black carbon,
- to determine the radiative forcing due to black carbon and
- to provide the datasets for climate model.

3. MATERIALS AND METHODS

3.1 **Site selection**

The proposed study will be carried out at Agra over IGB. Agra (27.17ºN, 78.0ºE) is located in the semi arid area of North Central India and situated at a distance of 200 km from the South-East of Delhi. Two-third of its peripheral border lines (SE, W and NW) are surrounded by the Thar Desert of Rajasthan. It represents one of the important sites over the Indo-Gangetic basin. The major industrial works are rubber processing, engineering, brick-kilns and chemical works. Besides, a number of petha industries are operating in the city. The vehicular emissions are one of the major sources of air pollution affecting the urban population in Agra. Unlike industrial emissions, vehicular pollutants are released at ground level and hence the impact on the recipient population will be more. The poor road, congested traffic and vehicular emission resulted into high pollution load.

The climate of Agra is dry, and it truly represents the subtropical region. Soil is sandy and loamy. Agra, situated on the Indo-Gangetic plain is about 169 m above the mean sea level (MSL) and has semiarid climate. The daily average temperature in summer lies between 21.9 $^{\circ}$C to 40 $^{\circ}$C and in winter temperature varies between 4.2 $^{\circ}$C to 31.7 $^{\circ}$C. The climate of Agra has been generally divided into four seasons: winter (December to February), summer (March to June), monsoon (July to September) and post monsoon (October to November). Most of the time atmosphere is in calm condition represents the wind velocity < 1 m s^{-1}. The wind follows two distinct patterns: during monsoon winds come from N, NE quadrant while in the rest of the season dominant winds are from NW sector.

3.1.1 Site description

In Agra, the sampling will be carried out at Dayalbagh, a suburban site. It is 10 km away from the industrial sector of the city where due to agricultural practices, vegetation predominates.

At Dayalbagh, all samples will be collected on the roof of a building in the Institute campus. The surrounding area is completely free of constructions and tall trees. Population around the sampling site is about 15,000. The soil is sandy and calcareous. A stockyard of about 1000 cattle is situated at a distance of 400 m west from the sampling site. The sampling site lies at a distance of about 200 m away from the side of the road that carries mixed vehicular traffic of the order of 10^3 vehicles in a day and a national highway (NH-II) at a distance of 2 km south has dense vehicular traffic (10^5 vehicles in a day). The site is surrounded by a variety of deciduous trees and agricultural fields. Agriculture is the major activity and fields around the site are covered with a variety of crops in the winter. In the summer uncultivated fields lie barren and dry.

3.2 Measurements in Air

Black carbon, particulate matter, and dry deposition will be measured.

3.2.1 Black carbon mass concentration

Black carbon (BC) concentration measurements will be carried out using the New seven channels Aethalometer (Model AE33; Magee Scientific USA). Aethalometer collect aerosol particles on the filter continuously by drawing air containing aerosol through the filter tape at a flow rate of 2 LPM. BC concentration will be estimated every minute following an optical attenuation technique. Aethalometer used continuous filtrations through quartz filter and estimates optical absorption using principle of transmission (Hansen et al., 1984). It measures changes at 7 different wavelengths (370, 470, 520, 590, 660, 880 and 970 nm). It is a self sufficient and automatic instrument and reproduces the data spontaneously. Fresher aerosols closer to the combustion sources show a larger ‘spot loading effect’. Well-aged aerosol shows almost zero effect under atmospheric conditions of high chemical activity and oxidative processing. The effect is revealed statistically by processing data collected over a large number of tape advances,

representing many data points collected at loadings (attenuation values) ranging from zero to the preset maximum (Drinovac et al., 2015). AE-33, a seven channel Aethalometer is programmed in such a way that it does not need any correction and can directly be taken into consideration.

3.2.2 Dry deposition of black carbon

Dry deposition of black carbon on leaves of Ashok (*Polyalthia longifolia*) and Amaltas (*Cassia fistula*) plants will be measured using surface washing methods (Davidson and Wu, 1990; Kumar et al., 2003).

3.2.2.1 Dry deposition on leaves

Dry deposition of black carbon samples will be collected from four leaves of Ashok (*Polyalthia longifolia*) and Amaltas (*Cassia fistula*). The leaves of selected plants will be tagged and washed with deionized water / chloroform using a sprayer for a number of times. The tagged and washed leaves will be air dried and exposed for 72 or 96 hrs on non rainy / non dewy / non foggy days. The black carbon deposit on the leave surface will be washed off into a beaker using 50 ml deionized water or chloroform and will be transferred to polypropylene bottles. The extracted BC particle will be filtered through a quartz filter.

Field blanks will be collected in the same manner as dry deposition on leaf surfaces by exposing the leaves for one minute.

3.2.3 Black carbon in soil

A representative number of soil samples will be collected from nearby places of the sampling site. Top of the soil will be removed and soil below the 1-2 cm will be collected. 1 gm of dried and sieved soil will be taken in 250 ml of conical flask and will be extracted in 100 ml E-pure water on electrical shaker for 2 hours. Extract will be filtered through pre-weighed quartz fiber

filter. The quartz fiber filter will be dried and weighed. The concentration of BC on quartz filter will be analyzed as the dry deposition sample.

3.3 Mass concentration of particulate matter

3.3.1 Sample collection, preparation and storage

Respirable Suspended Particulate Matter (PM_{10}) and fine particulate matter ($PM_{2.5}$) samples will be collected using the Polltech fine dust sampler at a flow rate of 16 LPM for 24 hrs using a glass fiber filter and PTFE filter, respectively. Pre-desiccated and pre weighed filter papers will be used as collecting surface and will be mounted in the filter holder and the instrument will be run for 24hrs. After the sampling filter will be withdrawn and will be kept in desiccators to remove the moisture and weight will be taken. Particle deposited on the filter will be determined gravimetrically. The difference in the mass of filter paper after the sampling and before the sampling will give loads of aerosol particles.

Half of the each filter will be cut into small pieces and extracted in 50 ml dionized water followed by half an hour extraction on ultrasonic bath. Extract will be filtered through Whatman 42 filter paper into polypropylene bottles. The sample will be stored in refrigerator till the analysis. The sample will be analyzed for major cations (Na^+, K^+, Ca^{2+}, Mg^{2+} and NH_4^+) and major anions (F^-, Cl^-, NO_3^-, SO_4^{2-}). All the plastic wares and glass wares will be cleaned by soaking in detergent for a few hours, followed by rinsing with deionized water for several times till conductivity of water reaches 1 µs and then will used for collecting/storing samples.

3.3.2 Analytical procedure

3.3.2.1 Analysis of BC in dry deposition and soil

Black carbon in dry deposition sample will analyzed using OC/EC analyzer.

3.3.2.2 Chemical Analysis

A representative number of samples of PM_{10} and $PM_{2.5}$ will be analyzed for major anions and major cations using Ion chromatograph (Dionex DX 1000) and Atomic Absorption Spectrophotometer.

3.4 Measurement of meteorological parameters

Air pollution status in any air shed is governed by meteorological conditions. Meteorological parameters viz., temperature, relative humidity, wind speed, wind direction and solar radiation will be monitored using a self contained battery operated AWS 10.0 Data logger (Ms/s Rave Innovations, Noida). AWS is a microprocessor based wind recording system with built-in clock and calendar. It consists of sensors for wind speed, wind direction, temperature, relative humidity and solar radiation. The data logger acquires data from the sensors for wind speed, wind direction, air temperature and solar radiation and stores the data in its memory for later retrieval as well as display the data to monitor. The global radiation will be monitored by using Pyranometer (Delta ohm). These global radiation lies in the range of 300-2800nm.

3.5 Estimation of radiative forcing

3.5.1 Optical properties of aerosol

The physical and optical properties of aerosols viz., aerosol optical depth, scattering and extinction coefficients, single scattering albedo, asymmetry factor measurements are cumbersome as they are highly variable over scale from local to regional to global and vary with location on earth and the season and very important in the radiation budget. Theoretical calculation of optical properties of aerosol will be carried out using the OPAC model (Hess et al., 1998).The new mixture of aerosol type will be defined in the OPAC model (Hess et al., 19998) software package and number densities of five components viz., insoluble, water soluble, soot, mineral accumulation

and mineral coarse will be incorporated to derive the aerosol optical parameters viz., aerosol optical depth (AOD), single scattering albedo (SSA), asymmetric parameter (ASP), extinction coefficient, absorption coefficient, scattering coefficient and angstrom coefficients for Agra region. Water soluble chemical constituents and minerals of aerosol measured during the study period will be used as input in OPAC.

3.5.2 Radiative forcing estimation

The aerosol radiative forcing due to BC represents the difference between the aerosol radiative forcing for aerosol with and without the BC component. The shortwave aerosol radiative forcing due to BC will be estimated using Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART) model (Ricchiazzi et al., 1998). SBDART is a FORTRAN computer code designed for the analysis of a wide variety of radiative transfer. The inputs values of SBDART model will be deduced from Optical Properties of Aerosols and Clouds (OPAC) model (Hess et al., 1998). The values of spectral aerosol optical depth and BC mass concentration will be measured and optical parameters for composites total aerosols will be deduced. In order to estimate the forcing due to BC alone, optical parameters for aerosols will be deduce again, keeping BC concentration equal to zero and constructed columnar AOD for BC free atmosphere from OPAC. However, the removal of BC increases the SSA value to a greater extent, which actually participate in aerosol radiative forcing (Gogoi et al., 2009; Morthy et al., 2010; Nair et al., 2014). In the present study in the SBDART model, tropical model atmospheric profiles will be used for aerosol and for aerosol free conditions. As Agra is on sand dunes surface reflectance properties of sand will be considered in the model calculation. The composite direct aerosol radiative forcing at the surface and the top of the atmosphere (TOA) will be estimated by calculating the differences in net shortwave radiative fluxes with and without black carbon aerosols. The flux in the atmosphere will be obtained by subtracting the flux at the TOA and at the surface.

3.6 Statistical Analysis

The data will be subjected for statistical analysis viz., mean, standard deviation, ANOVA test, correlation analysis, regression analysis, factor analysis using SPSS Vers. +6.0 software packages. The ANOVA test will performed to see the significance in the variation in the mean; correlation and regression analysis to the dependencies on variables and factor analysis to identify possible sources. ANOVA analysis will be used to see whether the difference in mean values of variables is significant between sites, seasons and collecting surfaces.

4. REFERENCS

Alam, K., Trautmann, T. and Blaschke, T (2011). Aerosol optical properties and radiative forcing over mega-city Karachi. Atmospheric Research 101, 773–782.

Allen, G. A., Lawrence, J. and Koutrakis, P. (1999). Field validation of a semi-continuous method for aerosol black carbon (Aethalometer) and temporal patterns of summertime hourly black carbon measurements in southwestern PA. Atmospheric Environment 33, 817– 823.

Babu, S.S., Satheesh, S.K., Moorthy, K.K. (2002). Aerosol radiative forcing due to enhanced black carbon at an urban site in India. Geophysical Research Letter 29 (18),1880.

Badarinath et al. (2007). Variation in black carbon aerosol, carbon monoxide and ozone over an urban area of Hyderabad, India, during the forest fire season. Atmospheric Research 85, 18–26.

Bano, T., Singh, S., Gupta, N.C., Soni, K., Tanwar, R.S., Nath, S., Arya, B.C. and Gera, B.S. (2011). Variation in Aerosol Black Carbon Concentration and Its Emission Estimates at the Mega-city Delhi. International Journal of Remote Sensing, 32 6749–6764.

Bergin et al. (2014). The discoloration of the Taj Mahal due to particulate carbon and dust deposition. Environmental Science and Technology. DOI: 10.1021/es504005q.

Bhugwant, C., Cachier, H., Bessafi, M. and Leveau, J. (2000). Impact of traffic on black carbon aerosol concentration at la Re´union Island (Southern Indian Ocean). Atmospheric Environment 24, 3463– 3473.

Bodhaine, B. A. (1995). Aerosol absorption measurements at Barrow, Mauna Loa and the south pole. Journal of Geophysical Research 100, 8967– 8975.

Bond et al. (2007). Historical emissions of black and organic carbon aerosol from energy-related combustion, 1850–2000. Global Biogeochemical Cycles 21, GB2018.

Bond, et al. (2013). Bounding the role of black carbon in the climate system: A scientific assessment. Journal of Geophysics Research 118, 5380–5552.

Brook, et al. (2010). Particulate matter air pollution and cardiovascular disease: an update to the scientific statement from the American Heart Association. Circulation 121, 2331–2378.

Bycenkiene, S., Ulevicius, V., and Kecorius, S. (2011). Characteristics of ˇ black carbon aerosol mass concentration over the East Baltic region from two-year measurements. Journal of Environmental Monitoring 13, 1027-1038.

Cao et al. (2006). Characterization of roadside fine particulate carbon and its 8 fractions in Hong Kong. Aerosol Air Quality Research 6 (2), 106–122.

Cao et al. (2014). Response of ocean acidification to a gradual increase and decrease of atmospheric CO_2. Environment Research Letter 9. doi:10.1088/1748-9326/9/2/024012.

Chen et al. (2014). Column aerosol optical properties and aerosol radiative forcing during a serious haze-fog month over North China Plain in 2013 based on ground-based Sunphotometer measurements. Atmospheric Chemistry and Physics14 (4), 2125–2138.

Chowdhury, Z., Hughes, L. S., Salmon, L. G. and Cass, G. R. (2001). Atmospheric particle size and composition measurements to support light extinction calculations over the Indian Ocean. Journal of Geophysical Research 106, 28,597–28,606.

Clarke, A. and Noone, K. (1985). Soot in the Arctic: a cause for perturbation in radiative transfer. Journal of Geophysical Research 19, 2045-2053.

Das, S.K and Jayaraman, A. (2011). Role of black carbon in aerosol properties and radiative forcing over western India during pre-monsoon period. Atmospheric Research 102, 320–334.

Dey, S., Sarkar, S., Singh, R.P. (2004). Comparison of aerosol radiative forcing over the Arabian Sea and the Bay of Bengal. Advances in Space Research 33, 1104–1108.

Dons et al. (2011). Impact of time-activity patterns on personal exposure to black carbon. Atmospheric Environment 45, 3594-3602.

Drinovec, et al. (2015). The “dual-spot” Aethalometer: an improved measurement of aerosol black carbon with real-time loading compensation. Atmospheric Measurement Techniques8, 1965–1979.

Gadhavi et al. (2015). Evaluation of black carbon emission inventories using a Lagrangian dispersion model -a case study over southern India. Atmospheric Chemistry and Physics 15, 1447–146.

Ginot et al. (2014). A 10 year record of black carbon and dust from a Mera Peak ice core (Nepal): variability and potential impact on melting of Himalayan glaciers. Cryosphere 8, 1479-1496.

Habib, G., Venkataraman, C., Chiapello, I., Ramachandran, S., Boucher, O., Reddy, M.S. (2006). Seasonal and interannual variability in absorbing aerosols over India derived TOMS: Relationship to regional meteorology and emissions. Atmospheric Environment 40, 1909-1921.

Hadley, O. L., Corrigan, C. E., Kirchstetter, T. W., Cliff, S. S. and Ramanathan, V. (2010). Measured black carbon deposition on the Sierra Nevada snow pack and implication for snow pack retreat. Atmospheric Chemistry and Physics10, 7505-7513.

Hansen, A.D.A., Rosen, H., Novakov, T. (1984). "The Aethalometer: an instrument for real-time measurement of optical absorption by aerosol particles". Science of the Total Environment 36, 191–196.

Hansen, J. and Nazarenko, L. (2004). Soot climate forcing via snow and ice albedo. Proceeding of National Academy of Science (USA) 101, 423-428.

Huang et al. (2014). High secondary aerosol contribution to particulate pollution during haze events in China. Nature 514, 218–222.

He et al. (2001). The characteristics of $PM_{2.5}$ in Beijing, China. Atmospheric Environment 35, 4959-4970.

Herich, H., Hueglin, C., Buchmann, B. (2011). A 2.5 year's source apportionment study of black carbon from wood burning and fossil fuel combustion at urban and rural sites in Switzerland. Atmospheric Measurement Techniques 4, 1409–1420.

Hess, M., P. Koepke, and I. Schult, 1998: Optical properties of aerosols and clouds: The software package OPAC. American Meteorological Society79, 831–844.

Hienola et al. (2013). Black carbon concentration and deposition estimations in Finland by the regional aerosol climate model REMO-HAMA. Atmospheric Chemistry and Physics 13, 4033-4055.

Im, J.S., Saxena, V. K. and Wenny, B. N. (2001). Temporal trends of black carbon concentrations and regional climate forcing in the southeastern United States. Atmospheric Environment 35, 3293– 3302.

Jacobson, M. Z. (2001). Strong radiative heating due to the mixing state of black carbon in atmospheric aerosols. Nature, 409, 695 – 697.

Jacobson, M. Z. (2004). Climate response of fossil fuel and biofuel soot, accounting for soot's feedback to snow and sea ice albedo and emissivity. Journal of Geophysical Research 109, D21201.doi:10.1029/2004JD004945.

Jayaraman, A., Gadhavi, H., Ganguly, D., Misra, A., Ramachandran, S., and Rajesh, T. A. (2006). Spatial variations in aerosol characteristics and regional radiative forcing over India: Measurements and modeling of 2004 road campaign experiment. Atmospheric Environment 40, 6504-6515.

Jian, W., Weimei, J. and Congbin, F. (2004). Simulation of the radiative effect of black carbon aerosols and the regional climate response over China. Advances in Atmospheric Sciences 21, 637-649.

Joshi et al. (2015). Investigations of aerosol black carbon from a semi-urban site in the Indo-Gangetic Plain region. Atmospheric Environment 125, 346-359.

Kompalli, S.K., Babu, S.S., Bharatan, L.N. and Moorthy, K.K. (2016). Spring-time enhancement in aerosol burden over a high altitude location (4520 m amsl) in western trans-Himalayas: results from long-term observations. Current Science 111, 117-131.

Kant, Y., Patel, P., Mishra, A.K., Dumka, U.C. and Dadhwal, V.K. (2012). Diurnal and seasonal aerosol optical depth and black carbon in the Shiwalik hills of the north western Himalayas: a case study of the Doon valley, India. International Journal of Geology, Earth and Environmental Sciences 2 (2).

Kanaya et al. (2016). Long-term observations of black carbon mass concentrations at Fukue Island, western Japan, during 2009–2015: constraining wet removal rates and emission strengths from East Asia.Atmospheric Chemistry And Physics16, 10689-10705.

Kaskaoutis, D.G., Sinha, P.R., Vinoj, V., Kosmopoulos, P.G., Tripathi, S.N., Misra, A., Sharma, M. and Singh, R.P. (2013). Aerosol properties and radiative forcing over Kanpur during severe aerosol loading conditions. Atmospheric Environment 79, 7-19.

Kedia, S., Ramachandran, S., Holben, B.N., Tripathi, S.N., (2014). Quantification of aerosol type, and sources of aerosols over the Indo-Gangetic Plain. Atmospheric Environment 98, 607-619.

Kim, Y. P., Moon, S.G., Lee, J. H. and Baik, N. J. (1999). Concentrations of carbonaceous species in particles at Seoul and Cheju in Korea. Atmospheric Environment 33, 2751–2758.

Kompalli et al. (2014). Aerosol black carbon characteristics over Central India: temporal variation and its dependence on mixed layer height. Atmospheric Research, 147 (8), 27–37.

Koch and Hansen (2005). Distant origins of Arctic black carbon: A Goddard Institute for Space Studies ModelE experiment. Journal of Geophysical Research 110, D04204.

Kompalli, S.K., Babu, S.S., Moorthy, K.K., Nair, V.S., Gogoi, M.M., Chaubey, J.P. (2013). Seasonal variation in the spatial distribution of aerosol black carbon over Bay of Bengal: A synthesis of multi-campaign measurements. Atmospheric Environment 64, 366-373.

Kumar, R., Rani, R., Singh, S.P, Satsangi, G.S., Kumari, K.M. and Srivastava, S.S. (2002). Measurement of dry deposition of gaseous and particulate S to marble at a semiarid region of India. Indian Journal of Radio and Space Physics 31, 88-92.

Kumar, R., Rani, A., Kumari, K.M. and Srivastava, S.S. (2005). Atmospheric dry deposition to marble and red stone. Journal of Atmospheric Chemistry, 50 (3), 253-271.

Kumar, R., Kumari, M.K. and Srivastava, S.S. (2006).Field measurements of aerosol particle dry deposition on tropical foliage at an urban site. Environmental Science and Technology 40, 135-14.

Kumar, R., and Kumari, K.M. (2015). Aerosols and trace gases characterization over Indo-Gangetic basin in semiarid region. Urban Climate 12, 11-20.

Latha, K. M. and Badarinath, K. V. S. (2003). Black carbon aerosols over tropical urban environment - A case study, Atmospheric Research 69, 125-133.

Latha, K.M. and Badarinath, K.V.S. (2005). Seasonal Variations of Black Carbon Aerosols and Total Aerosol Mass Concentrations over Urban Environment in India. Atmospheric Environment 39, 4129-4141.

Latha, R., Murthy, B.S., Lipi, K., Srivastava, M. K., Kumar, M., (2016). Absorbing Aerosols, Possible Implication to Crop Yield - A Comparison between IGB Stations. Aerosol and Air Quality Research 1-13.

Lavanchy et al. (1999). Elemental carbon (EC) and black carbon (BC) measurements with a thermal method and an Aethalometer at the high-alpine research station Jungraujoch, Atmospheric Environment 33, 2759-2769.

Leon et al. (2001). Large-scale advection of continental aerosols during INDOEX, Journal of Geophysical Research 106, 427-439.

Li et al. (2016). Sources of black carbon to the Himalayan–Tibetan Plateau glaciers. Nature Communication. DOI: 10.1038/ncomms12574.

Mahapatra et al. (2013). Variation in black carbon mass concentration over an urban site in the eastern coastal plains of the Indian sub-continent. Theoretical and Applied Climatology. DOI 10.1007/s00704-013-0984-z.

Moorthy, K. K., Babu, S. S., Kumar, S. V. S., Gupta, P. K., and Gera, B. S. (2004). Altitude profiles of aerosol BC, derived from aircraft measurements over an inland urban location in India. Geophysical Research Letter 31, L22103.

Nair, V.S., Babu, S.S., Moorthy, K.K., Sharma. A.K., Marinoni, A. and Ajai. (2013). Black Carbon Aerosols over the Himalayas: Direct and Surface Albedo Forcing. Tellus 65, 19738.

Núñez, X.C., Ruiz, L.V. and García, C.G. (2014). Black carbon and organic carbon emissions from wildfires in Mexico. Atmosphere 27, 165-172.

Paliwal, U., Sharma, M., Burkhart, J.F. (2016). Monthly and spatially resolved black carbon emission inventory of India: uncertainty analysis. Atmospheric Chemistry and Physics 16, 12457-12476.

Panicker, A. S., Pandithurai, G., Safai, P.D. and Kewat, S. (2008). Observations of enhanced aerosol long wave radiative forcing over an urban environment. Geophysical Research Letters 35, L04817.

Panicker, A.S., Pandithurai, G., Safai, P.D., Dipu, S. and Lee, D. (2010). On the contribution of black carbon to the composite aerosol radiative forcing over an urban environment. Atmospheric Environment 44, 3066-3070.

Patel, P.N. and Kumar, R. (2015). Estimation of Aerosol Characteristics and Radiative Forcing during Dust Events over Dehradun. Aerosol and Air Quality Research 15, 2082-2093.

Pathak, B., Kalita, G., Bhuyan, K., Bhuyan, P. K. and Moorthy, K.K. (2010). Aerosol temporal characteristics and its impact on shortwave radiative forcing at a location in the northeast of India. Journal of Geophysical Research 115, D19204.

Porch, W.M. and McCracken, M.C. (1982). Parametric study of the effects of Arctic soot on solar radiation. Atmospheric Environment 16, 1365-1371.

Raju, M.P., Safai, P.D., Sonbawne, S.M., Naidu, C.V. (2015). Black carbon radiative forcing over the Indian Arctic station, Himadri during the Arctic Summer of 2012. Atmospheric Research 157, 29-36.

Ram, K., Sarin, M. M. and Hegde, P. (2010). Long-term record of aerosol optical properties and chemical composition from a high-altitude site (Manora Peak) in Central Himalaya. Atmospheric Chemistry and Physics 10, 11791-11803.

Ramanathan, V. and Carmichael, G. (2008). Global and regional climate changes due to black carbon. Nature Geosciences 1, 221-227.

Raatikainen, T., Brus, D., Hyvärinen, A.P., Svensson, J. Asmi, E. and Lihavainen, H. (2015). Black carbon concentrations and mixing state in the Finnish Arctic. Atmospheric Chemistry and Physics 15, 10057-10070.

Reddy, B.S.K., Kumar, K.R., Balakrishnaiah, G., Gopal, K.R., Reddy, R.R., Reddy, L.S.S., Ahmmed, Y.N., Narasimhulu, K. and Moorthy, K.K. (2012). Potential Source Regions Contributing to Seasonal Variation of Black Carbon Aerosols over Anantapur in Southeast India. Aerosol and Air Quality Research 12, 344-358.

Rehman, I.H., Ahmed, T., Praveen, P.S., Kar, A. and Ramanathan, V. (2011). Black carbon emissions from biomass and fossil fuels in rural India. Atmospheric Chemistry and Physics 11, 7289–7299.

Rosen, H. and Novakov, T. (1977). Raman scattering and the characterization of atmospheric aerosol particles. Nature 266, 708-710.

Rosen, H., Novakov, T. and Bodhaine, B. (1981). Soot in the Arctic. Atmospheric Environment 15, 1371-1374.

Ruellan, S., and Cachier, H. (2001). Characterization of fresh particulate vehicular exhausts near a Paris high flow road. Atmospheric Environment 35, 368-453.

Ruppel et al. (2013). Comparison of Spheroidal Carbonaceous Particle Data with Modelled Atmospheric Black Carbon Concentration and Deposition and Air Mass Sources in Northern Europe, 1850–2010. Advances in Meteorology 15, 393926.

Safai, P.D., Kewat, S., Praveen, P.S., Rao, P.S.P., Momin, G.A., Ali, K. and Devara, P.C.S. (2007). Seasonal variation of black carbon aerosols over a tropical urban city of Pune, India. Atmospheric Environment 41, 2699– 2709.

Safai, P.D., Raju, M.P., Maheshkumar, R.S., Kulkarni, J.R., Rao, P.S.P., Devara, P.C.S. (2012). Vertical profiles of black carbon aerosols over the urban locations in South India. Science of the Total Environment 431, 323–331.

Sarkar, C., Chatterjee, A., Singh, A.K., Ghosh, S.K., Raha, S. (2015). Characterization of Black Carbon Aerosols over Darjeeling-A High Altitude Himalayan Station in Eastern India. Aerosol and Air Quality Research 15, 465-478.

Saud et al. (2012). Emission estimates of organic and elemental carbon from household biomass fuel used over the Indo-Gangetic Plain (IGP). Atmospheric Environment 61, 212-220.

Sharma, S., Ishizawa, M., Chan, D., Lavoué, D., Andrews, E., Eleftheriadis, K., and Maksyutov, S. (2013). 16-year simulation of Arctic black carbon: transport, source contribution, and sensitivity analysis on deposition. Journal of Geophysical Research 118, 1-22.

Singh, A., Rajput, P., Sharma, D., Sarin, M. M. and Singh, D. (2014). Black Carbon and Elemental Carbon from Postharvest Agricultural-Waste Burning Emissions in the Indo-Gangetic Plain. Hindawi Publishing Corporation Advances in Meteorology 179301, 10.

Srivastava, A.K., Tripathi, S.N., Dey, S., Kanawade, V.P., Tiwari, S. (2012). Inferring aerosol types over the Indo-Gangetic Basin from ground based sunphotometer measurements. Atmospheric Research 109-110, 64–75.

Streets et al., (2003). An inventory of gaseous and primary aerosol emissions in Asia in the year 2000. Journal of Geophysical Research 108, 8809. doi:10.1029/2002JD003093.

Sullivan, A. P., Weber, R. J., Clements, A. L., Turner, J. R., Bae, M. S. and Schauer, J. J. (2004). A method for on-line measurement of water soluble organic carbon in ambient aerosol particles: results from an urban site. Geophysical Research Letter 31, L13105.

Suman, D. (2013). Dry Deposition Removal of Carbonaceous Particles Produced by Biomass Burning in Panama. Aerosol Science and Technology, 1521-7388.

Surendran, D.E., Beig, G., Ghude, S.D., Panicker, A. S., Manoj, Dilip, M. G., Chate, M and Ali, K. (2013). Radiative forcing of black carbon over Delhi. International Journal of Photoenergy 313652.

Takeda et al. (2012). Optical method for measuring deposition amount of lack carbon particles on foliar surface. Asian Journal of Atmospheric Environment 6-4, 268-274.

Thompson, L.G. (2003). Tropical glacier and ice core evidence of climate changes on annual to millennial time scales. Climatic Change 59, 137-155.

Tiwari, S., Srivastava, A.K. Bisht, D.S., Parmita, P., Srivastava, M.K., Attri, S.D. (2013). Diurnal and seasonal variations of black carbon and $PM_{2.5}$ over New Delhi, India: Influence of meteorology. Atmospheric Research 125-126, 50-62.

Tiwari, S., Srivastava, A.K., Bisht, D.S., Bano, T., Singh, S., Behura, S., Srivastava, M.K., Chate., D.M., Padmanabhamurty, B. (2009). Black carbon and chemical characteristics of PM_{10} and $PM_{2.5}$ at an urban site of North India. Journal of Atmospheric Chemistry 62 (3), 193-209.

Tripathi, S. N., Dey, S. and Tare, V. (2005). Aerosol black carbon radiative forcing at an industrial city in northern India. Geophysical Research Letters 32, L08802.

USEPA. (2012). Black carbon effect on public health and environment. Department of the Interior Environment and Related Agencies Appropriation Act, 2010. EPA-450/R-12-001.

Vaishya, A., Singh, P., Rastogi, S., Babu, S.S. (2016). Aerosol black carbon quantification in the central Indo-Gangetic Plain: Seasonal heterogeneity and source apportionment. Atmospheric Research 185, 13-21.

Venkataraman, C., Habib, G., Eiguren- Fernandez, A.L., Miguuel, A.H., Friendlanden, S.K. (2005). Residential Bio-fuel in South Asia: Carbonaceous Aerosol Emissions and Climate Impacts. Science 307, 1454-1456.

Venkataraman, C., Reddy, C. K., Josson, S. and Reddy, M. S. (2002). Aerosol size and chemical characteristics at Mumbai, India during the INDOEX-IFP (1999). Atmospheric Environment 36, 1979-1991.

Verma, S., Venkataraman, C. and Boucher, O. (2011). Attribution of aerosol radiative forcing over India during the winter monsoon to emissions from source categories and geographical regions. Atmospheric Environment 45, 4398-4407.

Wang et al. (2011). Sources of carbonaceous aerosols and deposited black carbon in the Arctic in winter-spring: Implications for radiative forcing. Atmospheric Chemistry and Physics 11 (23), 453-473.

Wang et al. (2014). Exposure to ambient black carbon derived from a unique inventory and high-resolution model. Proceedings of the National Academy of Sciences, USA 111, 2459-63.

WHO, 2014. World Health Organization: 2014 Air Pollution Ranking. http:faq/2015-05-16/world-health-organization-2014-air-pollution-ranking.

Yasunari et al.(2010). Estimated impact of black carbon deposition during pre-monsoon season from Nepal Climate Observatory-Pyramid data and snow albedo changes over Himalayan glaciers. Atmospheric Chemistry and Physics 10 (14), 6603-6615.

Yasunari et al. (2013). Estimated range of black carbon dry deposition and the related snow albedo reduction over Himalayan glaciers during dry pre-monsoon periods. Atmospheric Environment 78, 259-267.

Yu, A., Zenderb, C.S., Saxena, V.K. (2001). Direct radiative forcing and atmospheric absorption by boundary layer aerosols in the southeastern US: model estimates on the basis of new observations. Atmospheric Environment 35, 3967-3977.

Zhang, H. and Wang, Z. (2011). Advances in the study of black carbon effects on climate**.** Advances in Climate Change Research 2, 23-30.